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BUILDING A POLLUTANT RESERVOIR ALOFT BY CUMULUS CLOUDS

by

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16. ABSTRACT

A diagnostic analyses is presented to assess the impact of non-precipitating clouds on rates of change of pollutant concentration both in the mixed layer and in the free troposphere. This analyses is based on a coupled two layer (mixed and free tropospheric) conservation of pollutant species model which distinguishes the role of active or venting type clouds from the forced, or fair weather cumulus clouds. The study examines the sensitivity of the modeled rates of change of pollutant predictions in the mixed layer and in the free troposphere to the method by which the vertical exchange of these venting class clouds is modeled. This analyses focus on the vertical redistribution process, the heterogeneous conversion process which are known to be rapid and important is ignored in this study by considering only total pollutant concentration. Details of the processes at the top of the mixed layer entrainment zone is highly simplified in this model.

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1. INTRODUCTION

Cloud venting refers to the vertical transport of mixed layer pollutants into the overlying cloud layer by cumulus convective clouds (Ching, 1982). Precipitating clouds deposit a fraction of their pollutant burden to the surface. Other cumulus clouds and the remnants of these clouds may store the residual pollutants in situ, thereby creating a reservoir of pollutants aloft (Ching et al. 1984). The horizontal extent of non precipitating convective cloud activity is highly variable both spatially and temporally and is, therefore, apt to contribute to the variability in pollutant concentration in both mixed and cloud layers. Thus, the modeling of the transport and transformation processes associated with cumulus cloud activity is an important consideration for the regional scale air pollution models.

The development of parametric models for nonprecipitating cloud processes requires an understanding of the interaction between the meteorological environment and the cloud fields. Mixed layer pollutants condensing within rising thermals undergo rapid chemical transformation in the clouds. Upon evaporation, these compounds either remain within the mixed layer or add to the pollutant reservoir above the mixed layer depending on the vertical extent of the clouds (Stull, 1985).

A diagnostic model is presented to assess the impact of non-precipitating cloud regimes on rates of change of pollutant concentration both in the mixed layer and in the cloud layer. This analysis is based on a coupled, two-layer (mixed and free tropospheric) conservation of pollutant species model which distinguishes the role of active or venting type clouds from the forced, or fair weather cumulus clouds. The study examines the sensitivity of the modeled rates of change of pollutant predictions in the mixed layer and in the free troposphere to the method by which the vertical exchange of these venting class clouds is modeled. This analysis focuses on the vertical redistribution process. The heterogeneous conversion process, which is known to be rapid and important, is ignored in this study by considering only total pollutant concentration.

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2. MODEL FORMULATION

The rate of change of mixed layer pollutant concentration in a grid volume, bounded by the surface and the top of the mixed layer, depends on the differential advection of the pollutant, the net production or destruction, the net deposition to the surface, and the net flux of the pollutant out of the mixed layer by cloud activity (Equation 1).

$$\int_0^h (\partial X/\partial t)dz = \int_0^h (ADV + P)dz - F_2 + F_1 \qquad (1)$$

where X = mixed layer concentration (ppb), ADV = advection component, P = net production or loss, F_2 = loss at h (top of the mixed layer) due to cloud venting and F_1 = deposition loss to the surface. The surface deposition is parameterized as the product of a deposition velocity, v_d, for the species times its concentration value. Strict specification of the deposition velocity approach requires a height dependency formulation. This is ignored in this diagnostic study.

Equation 2 refers to the pollutant concentration, Y, in the free troposphere or cloud layer whose vertical extent is the top of the mixed layer to the limit of free convection(h_c).

$$\int_h^{h_c} (\partial Y/\partial t)\, dz = F_2 \qquad (2)$$

Advection and production terms are neglected in (2).

There is no formulation currently available for computing the cloud flux. Heuristically, the exchange should be related to (a) the difference of concentration between the two layers and (b) a scaling velocity for transport, i.e., a "venting velocity", V_t, made up of fractional cloud coverage, a_c, and updraft strength, W_c.

$$F_2 = Ca_c W_c (X-Y) = V_t (X-Y) \qquad (3)$$

C is a coefficient that expresses the net effect of up and downdrafts in determining the vertical exchange rate. Downdrafts induced by clouds (Knupp and Cotton, 1985) are believed to play an important role in the vertical exchange process (Ching and Alkezweeny, 1985), and a realistic method by which they are treated in models is of critical importance to predicted exchange rates. Upward motion and the subse-

quent downward motions induced by venting clouds can be envisioned to occur between two extremes. In one extreme, the penetrative updrafts are viewed as a temporary state to be followed by subsequent downdrafts which return the cloud volume (chemically converted) back into the mixed layer; as a result, little or no net vertical transport occurs. In this case, C=0. On the other extreme, the maximum vertical exchange occurs when active clouds force free tropospheric air into the mixed layer concurrently as or soon after such clouds rise above the mixed layer. In this case, C=1. Thus, the details of cloud up and downdrafts require detailed knowledge of cloud entrainment processes which are not well understood to date. Aircraft measurements from EPA's Northeast Regional Oxidant Study (NEROS) of vertical flux measurements of ozone within and in the surrounding domain of individual venting clouds were made using a fast response, airborne three-dimensional wind (gust probe) system, and a fast response ozone monitor (Ching et al. 1983). Greennut (1986) analysed data taken during NEROS 1979 and 1980 and obtained for an ensemble of clouds,

$$C = 0.79 \qquad (4)$$

The venting velocity for this model is based on this result. This assumes the transport by the large-scale subsidence component of the downdraft induced by the rising cumulus elements to be negligible. When introducing this component into the analyses, Greenhut finds C = 1.12, a value greater than one which he attributes to cloud turbulence. Clearly, much more effort will be required to establish the validity of this parametric formulations. Closure for V_t must also include information on the areal extent of clouds within a grid area, the vertical distribution of convective clouds within the cloud field, the intensity of the up and downdrafts of each and all clouds in the grid, the type of meteorological environment between the surface and the limit of free convection, and of course, the source region for the induced downdrafts. In principle, closure regarding the cloud coverage a_c is possible using satellite data, weather observations, etc.

The formulation of cloud flux depends explicitly on the concentration of both the mixed and the cloud layer. Thus, (1) and (2) can be written as follows

$$DX = S - GX + F\,(Y-X) \qquad (5)$$

and

$$DY = F\,h\,(X-Y)/\lambda \qquad (6)$$

where $G = V_d/h$, $F = V_t/h$, λ is the thickness of the cloud layer, S is a source term which treats the advection and production component together, and D is the differential operator $\partial/\partial t$. These expressions assume the concentration within the mixed and cloud layers to be instantly homogenized within their respective layers. Further, the entrainment processes by dry convective motions at the top of the mixed layer are ignored. Setting the parameters to constants, (5) and (6) can be readily solved to yield exact solutions given by:

$$X = A(e^{m_1 t} - e^{m_2 t}) + B\,e^{m_2 t} + E \qquad (7)$$

and

$$FY = A(m_1 e^{m_1 t} - m_2 e^{m_2 t} + (F+G)\,(e^{m_1 t} - e^{m_2 t}) + H\,e^{m_2 t} + JE \qquad (8)$$

where

$$A = (FY_0 + S(m_2/G + 1) - X_0(m_2+F+G))/(m_1-m_2) \qquad (9a)$$
$$B = X_0 - C \qquad (9b)$$
$$E = S/G \qquad (9c)$$
$$H = B\,(m_2+F+G) \qquad (9d)$$
$$J = FE \qquad (9e)$$
$$b = 0.5(G+F(1+h/\lambda))_2 \qquad (9f)$$
$$m_1 = -b-0.5\ \mathrm{sqrt}\ (b^2-4hFG/\lambda) \qquad (9g)$$
$$m_2 = -b+0.5\ \mathrm{sqrt}\ (b^2-4hFG/\lambda) \qquad (9h)$$

X_0 and Y_0 are the initial concentrations. The requirement that the parameters and the source term be time invariant is an over simplification, but suitable for this diagnostic study. The cloud convection time period is only likely to occur between late morning and late afternoon, when the cloud activity, deposition, and production, mixed layer height and top of the cloud layer may exhibit some degree of stationarity. Other analytical solutions to (5) and (6) for more general situations are, in principle, obtainable.

3. RESULTS AND DISCUSSION

The major parameters for this two-layer model are S, G, F, X_0, Y_0, h, and λ. Figures 1 and 2 describe the time series of mixed layer and cloud layer concentrations for negligibly small and average value of S. In this survey, the deposition velocity was assumed to be 1 cm s^{-1}, X_0 = 50 ppb, Y_0 = 25 ppb, h and λ = 1 km. For diagnostic purposes, a reference or modal value for F was set to be the same as the ground loss rate, which corresponds to V_t = 1 cm s^{-1}. A value of V_t = 10 was chosen to represent large cloud flux. To illustrate, V_t = 1 would apply to cumulus updrafts of 1 m s^{-1} for an areal coverage of 1% of cumulus clouds. Similarly, 2 m s^{-1} updrafts covering 5% yields V_t = 10, etc.

Figure 1 show for source term of zero cases for a) negligible cloud flux, b) modal cloud flux, and c) relatively large cloud flux. Figure 1a shows that the mixed layer ozone decreases with time from its initial value of 50 ppb by about 30% after 10 hours due to surface dry deposition. Since cloud flux was negligible the concentration in the cloud layer remain unchanged. Figure 1b shows the mixed layer concentration to decrease by 37% after 10 hours with the addition of cloud venting, and the cloud layer ozone concentration to increase with time from 25 ppb to 29 ppb. Increasing the cloud transport as in Fig 1c decreases the mixed layer concentration by 40% and increases the cloud layer concentration to 32 ppb after 10 hours. Generally, the rate of decrease of X and the rate of increase of Y with time show a dependency on V_t. Interestingly, the cloud layer concentration exceeds and remains higher than that in the mixed layer

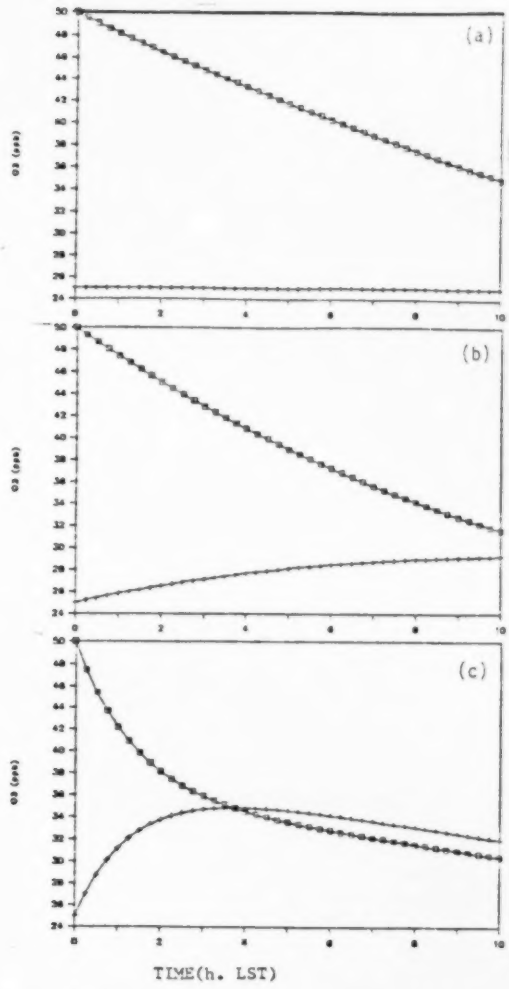


Figure 1. Time series of ozone concentration in the mixed layer (boxes), and for the cloud layer, (pluses) for negligible oxidant production, and for a, b, and c refering to zero, modal and moderately large cloud venting velocities.

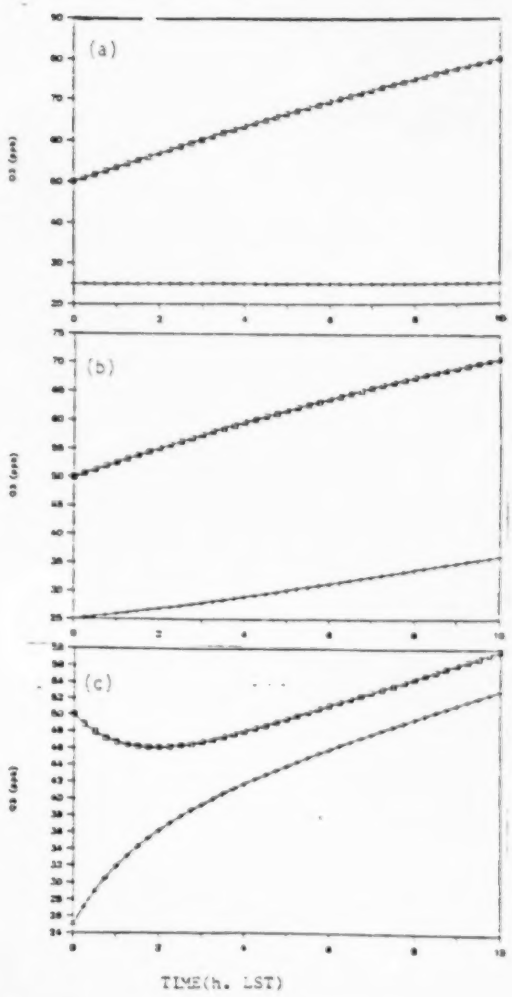
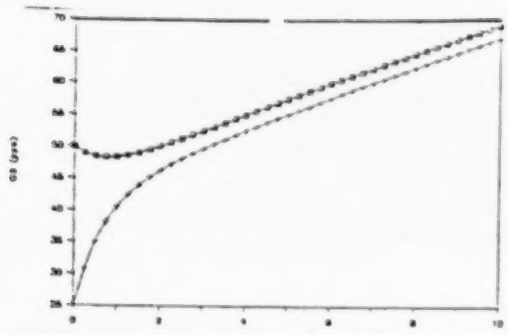


Figure 2. Same as Figure 1 except for source strength of 5.4 ppb h^{-1}.

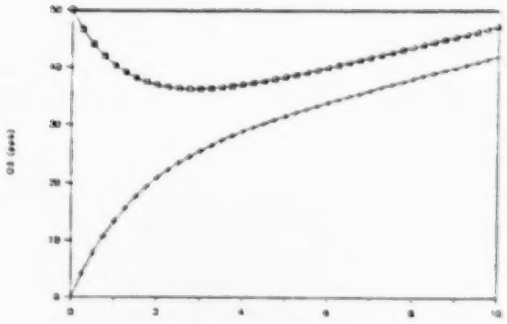
after 3.5 hours in Figure 1c. During this period, the cloud layer acts as a source region for the mixed layer. The time at which the cloud layer concentration becomes equivalent to or exceeds that in the mixed layer decreases with increasing magnitude of V_t from about 10 hours for V_t less than 1 cm s^{-1}, to 5 hours for V_t = 5 cm s^{-1} and to 2 hours for V_t = 20 cm s^{-1}.

Figure 2 refers to the results for a typical source value, S, of modal value 0.0015 ppb s^{-1} or 5.4 ppb h^{-1}. Figure 2a refers to the case of negligible cloud flux, and the production rate is sufficiently greater than dry deposition allowing the mixed layer to increase its concentration by 60% to 80 ppb. Here again, the concentration in the cloud layer is time invariant. Figure 2b refers to a cloud flux velocity equivalent to the dry deposition velocity. The mixed layer concentration increases to 70 ppb, a 40% increase and is less than that for the no cloud flux situation. The cloud layer concentration shows an increase from 25 ppb to 36 ppb, greater than that for the case in Figure 1b. Figure 2c refers to a moderately large cloud flux, 10 times that of the surface deposition values. Greenhut (1986) observed a mean flux velocity of 6.0 cm s^{-1} during NEROS. The mixed layer concentration actually decreases with time for the first 1.75 hours as convective cloud venting occurs. After 10 hours, the mixed layer ozone has increased by only 16%, smaller than that for the case of smaller cloud flux. Thereafter, the magnitude of the difference in the concentration of ozone between the mixed and cloud layer becomes sufficiently small that the production continues to increase the concentrations



TIME(h. LST)

Figure 3. Same as Figure 2c except that h/λ = 3.



TIME(h. LST)

Figure 4. Same as Figure 2c except that $Y_0/X_0=0$.

in both mixed and cloud layers. Thus, for increasing magnitudes of V_t the rate of increase of X decreases and the rate of increase of Y increases.

It can be shown that the solution for X and Y are dependent uniquely on h/λ. Additionally, after 10 hours, X and Y values are greater for larger values of h/λ. This can be seen when comparing model results of Figure 2c with those of Figure 3. Figure 3 model results are for h/λ = 3, while Fig. 2c is for h/λ = 1.

The relative value of the difference between the initial concentration of cloud layer ozone from that of the mixed layer affects the solution to this model. Figure 4 shows results for the case of Figure 2c with the exception that Y_0 = 0 rather than 25 ppb. Clearly, both mixed and cloud layer concentration values exhibit greater values after 10 hours when the ratio of Y_0 to X_0 increases.

4. SUMMARY AND CONCLUSIONS

A coupled mixed-cloud layer model provides a means to diagnose the changes in ozone concentration in both layers when vertical exchange by cloud venting processes augments dry deposition to the surface in modulating oxidant production and advection. Coefficients related to deposition, venting, production/advection are time invariant in this study. The formulation for the flux is the product of a venting velocity and the concentration difference between the mixed and cloud layers. The venting clouds are the active clouds defined by Stull(1985). His forced clouds are the fair weather cumulus which are restricted to the mixed layer and are not treated in detail in this study. The venting velocity parameterization incorporates cloud induced downdrafts implicitly in its formulation. The formulation of the venting velocity is based on empirical data collected from aircraft observations.

Sensitivity tests performed on the model show that active clouds can significantly impact ozone concentration levels in both the mixed and the cloud layers during the convective period. However, the range of cloud exchanges is large. The predicted mixed and cloud layer concentrations also vary with h/λ, and Y_0/X_0.

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